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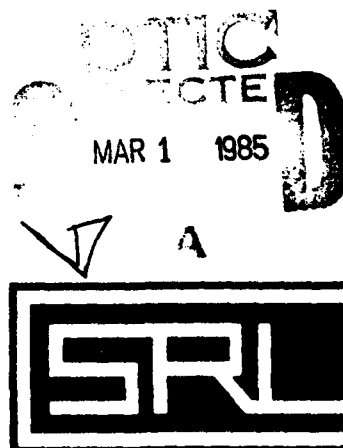
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Three rare-earth-ion crystals were shown to be good candidates for surface temperature measurements on energetic materials by laser-induced fluorescence. Dy^{+3}:LaF$_3$ displays a thermalization process with an energy gap of 1070/cm^{-1} (approx. energy level), resulting in a fluorescence intensity change of $\sim$ 200 over a 700-K temperature range. Cr^{+3}:Al$_2$O$_3$ (ruby) displays an extreme temperature sensitivity to the lifetime of the R-fluorescence lines. The lifetimes are observed to change by a factor of $\sim$ 230 over a 500-K temperature range. Er^{+3}:CaF$_2$ shows a temperature sensitivity of 20 over a temperature range of 700 K, as measured by the lifetime of its E(^{4}S$_{3/2}$) fluorescence line. Originator Supplied Keywords include:

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APPLICATION OF ATOMIC FLUORESCENCE TO MEASUREMENT OF COMBUSTION TEMPERATURE IN SOLID PROPELLANTS

Annual Progress Report on
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Prepared for

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December 1984

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LIST OF ILLUSTRATIONS

I. INTRODUCTION

The condensed-phase combustion processes of energetic materials in inter-
mediate (3-20 MPa) and high (> 300 MPa) pressure regions presents many
challenges for researchers in the field of optical diagnostics; to date,
this area has received less attention than the experimentally more tract-
able problem of gas-phase combustion. The objective of the present study
was to identify and develop optical diagnostic probes which can be used to
measure the surface temperature of an energetic material. The choice of a
diagnostic technique for surface-temperature measurements is complicated by
the requirements of high spatial resolution (1 μm), high temporal resolu-
tion (1 μs or less per sample time), nonintrusiveness, remote probing of
temperature, and making measurements on an ever-changing reacting surface.

Advanced optical diagnostic techniques such as coherent anti-Stokes Raman
spectroscopy (CARS), ordinary Raman spectroscopy, and laser-induced fluo-
rescence (LIF) have been employed extensively for combustion measurements
in atmospheric flames and plumes of solid propellants.[1-6] However, no at-
tempt has been made to apply these techniques to the measurement of react-
ing surface temperatures. Of the available optical diagnostic techniques,
LIF holds the most promise of meeting the criteria for an effective diag-
nostic method for surface-temperature measurements. However, to the
authors' knowledge, there is no molecular or atomic species naturally
existing in propellants which can be used for LIF surface-temperature meas-
urements. Thus, for this technique to be employed, a fluorescent species
must be introduced into the propellant or model propellant material whose

emission spectrum displays sensitivity to temperature change over the temperature range of interest.

Atomic species such as indium and thallium which have low-lying electronic states populated at relatively low temperatures have been successfully employed in atmospheric flames for thermometry.[5] However, because of the amorphous nature of propellants, the atomic fluorescence is either quenched or homogeneously broadened to the point where it cannot be detected. Attention was thus directed away from the conventional atomic species to species such as rare-earth ions which are known to fluoresce in the solid state and be quite sensitive to temperature changes. Since rare-earth ions are not conventional dopant species for thermometry, discussion of the rare-earth spectral sensitivity to temperature is in order.

Temperature may affect the spectrum of rare-earth ions in several ways. First, the addition of heat causes the crystal lattice containing the rare earth to vibrate which, in turn, creates a changing crystal field at the location of the rare-earth ion and produces a broadening in the linewidth of the optical transitions. Secondly, as the temperature is increased, the number of upper levels of the system which become populated increases according to Boltzmann's law: $\exp(-\Delta E/kT)$. This "thermalization" effect causes a change in the intensity distribution of the rare-earth spectrum with temperature. Thirdly, a frequency shift of the spectral lines can occur due to the thermal expansion of the crystal lattice. The greater average ion separation at high temperatures results in a reduced crystal

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field and, therefore, in reduced Stark-component separation. Finally, as the temperature increases, the vibration of the crystal lattice (phonons) can nonradiatively relax the excited electronic state, thus reducing the lifetime of the radiative transition (quenching).

Of the many temperature effects demonstrated by the rare-earth ions, thermalization and lifetimes are the most sensitive in the temperature range 300-1300 K.

II. OBJECTIVES

INTRODUCTION

The overall aim of this program is to study the surface reactions of a solid rocket propellant and to obtain temperature data which can be related to the kinetics of the combustion process. The technique of laser-induced fluorescence (LIF) is being utilized to measure the fluorescence spectrum and fluorescence lifetime of an impurity introduced into the reacting material. Since the fluorescence spectra and lifetime of the impurity are sensitively dependent upon temperature, it should be possible to obtain a temperature profile of the reacting surface.

The primary goal of the program is to demonstrate that laser-induced fluorescence measurements can be directly related spatially and temporally to the surface-temperature profile of a solid undergoing very rapid heating. This is accomplished by selecting a model material which closely simulates the reacting surface of a solid propellant and subjecting it to high-energy flux from a cw CO_2 laser.

EXPERIMENTAL APPROACH

In order to utilize laser-induced fluorescence (LIF) in this study, three experimental objectives must be accomplished. The first, which is fundamental to the fluorescence technique, involves selection of a proper dopant

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which can be incorporated into the reacting material and will possess sufficient fluorescence to yield surface temperatures. The second is the selection of a model material which, upon rapid heating, will simultate the surface of a reacting solid rocket propellant. The third objective is to show unequivocally that the temperatures inferred from the fluorescence measurements actually represent the surface temperature of the solid being studied under conditions of rapid (10^4 K/sec) heating.

Phase I

During this initial phase a fluorescence material and a matrix material to contain the fluorescent material will be selected, and preliminary spectroscopic data will be obtained. A test cell with optical access and provision for slow heating of the samples has been constructed to perform the testing. Final tests are being performed in preparation for Phase II.

Phase II

Once a proper medium for fluorescence measurements has been selected and specimen heating accomplished, verification of surface temperature and fluorescence temperature will be performed.

A high-speed-pyrometer system compatible with the test cell will be constructed.

Extensive measurements will be performed to correlate the surface tempera-
ture measured using the pyrometer system and the fluorescence technique
with thermocouple temperatures.

Phase III

Once the technique of laser-induced fluorescence (LIF) has been experimen-
tally verified, surface-temperature measurements under rapid heating con-
ditions will be initiated.

Both inert and reactive atmospheres will be utilized in these experiments,
and both LIF and pyrometry will be used to measure temperature.

Also, other matrix and fluorescence materials will be evaluated to assure
that a realiable, accurate technique has been developed for profiling the
temperature of a reacting surface.

Phase I has been completed within this first year of the program. Phase II
is currently underway. Personnel connected with the fulfillment of this
contract are as follows:

Co-Principal Investigator:	L. P. Goss, Ph.D.
Co-Principal Investigator:	A. A. Smith, Research Physicist
Physical Science Technician:	M. Rowe

III. EXPERIMENTAL SYSTEM

The experimental system employed for both the thermalization and the lifetime studies is shown in Fig. 1. The tripled output of a Molectron Nd:YAG laser (355 nm) was used to excite the test specimen. A focusing lens was used to restrict the pump-beam size on the doped crystal and increase the fluorescence to permit adequate detection. The fluorescence was collected and directed into a J-Y Model HRP 0.6-m monochromator where it was detected via an S-20 photomultiplier. Thermalization measurements were made by recording the fluorescence spectra over the frequency range of interest on a strip-chart recorder. The lifetime measurements were made with a Model 1800 Biomation Transient Digitizer which digitized the data from the fluorescence decay curve for storage and analysis on a Digital PDP Micro 11-23+ computer system. The lifetimes were determined by fitting a single exponential function and taking the lifetime as the 1/e value of that function.
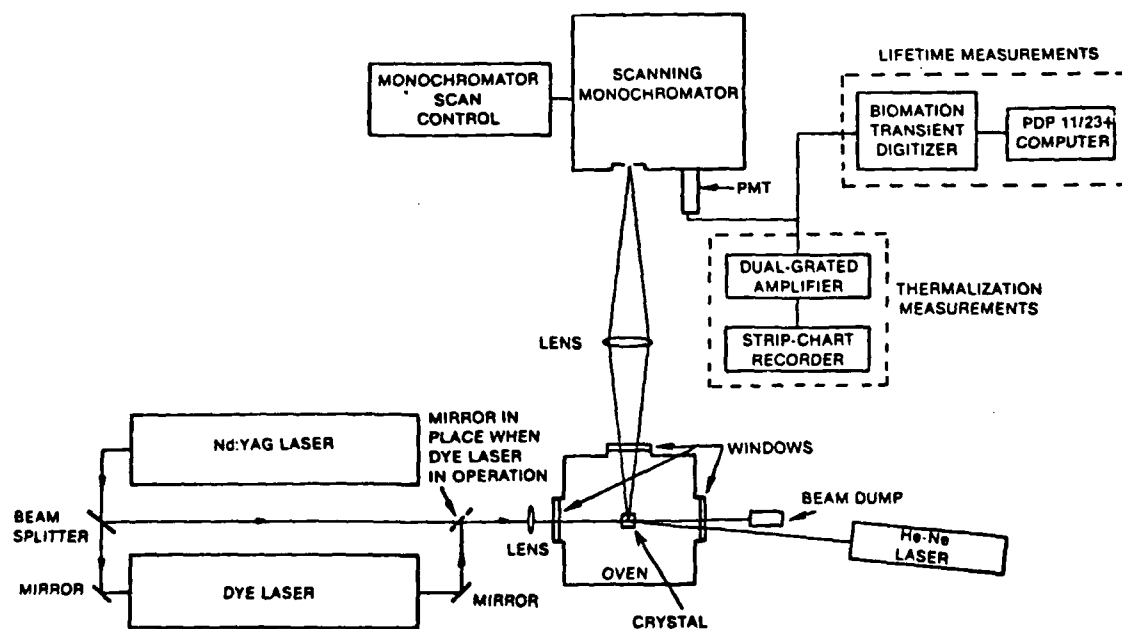
Figure 1. Experimental Setup for Thermalization and Lifetime Measurements.

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Three rare-earth crystals were chosen for study--$Cr^{+3}:Al_2O_3$, $Er^{+3}:CaF_2$, and $Dy^{+3}:LaF_3$. The $Er^{+3}:CaF_2$ and $Dy^{+3}:LaF_3$ crystals were supplied by Optovac, Inc. The ruby crystal was taken from a ruby rod used in a Korad ruby laser system.

The oven employed in these studies was a Nichrome coiled system encased in ceramic, capable of reaching temperatures as high as 1223 K. The oven was controlled by a West Model 2070 Digital Temperature Controller, with the temperature being measured by means of a Chromel-Alumel thermocouple.

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IV. RESULTS AND DISCUSSION

THERMALIZATION

When two rare-earth-ion energy levels are separated by less than 1000 cm^{-1}, the upper level typically will not fluoresce at low temperatures.[7] The reason for this behavior is that multi-phonon relaxation rates are extremely high (10^6/s) with small energy gaps since fewer phonons are required for quenching. Thus, at low temperatures there is no population build-up in the upper energy level and, therefore, no fluorescence is observed. However, as the temperature increases, the upper level becomes increasingly populated due to the very fast thermal equilibration among Stark levels. This rate of thermal equilibration is much faster than the radiative and nonradiative decay rates; thus, the upper-level population increases according to the Boltzmann Equilibrium Law. As the temperature increases, the population buildup in the upper level increases, and the fluorescence increases relative to the lower energy level.

An example of this behavior can be found in Dy^{+3}:LaF_3.[8] The important energy levels for this rare earth are shown in Fig. 2. The pump-excitation wavelength is 355 nm (tripled 1064 nm). The absorbed laser light excites Dy^{+3} in a high-energy level which radiatively and nonradiatively decays to the F($^4F_{9/2}$) level. This level undergoes a fast thermal equilibrium which pumps a portion of its population into the nearby G($^4I_{15/2}$) level. The fluorescence is then observed from both states. In Fig. 3 the experimentally observed G and F fluorescence from Dy^{+3} as a function of temperature
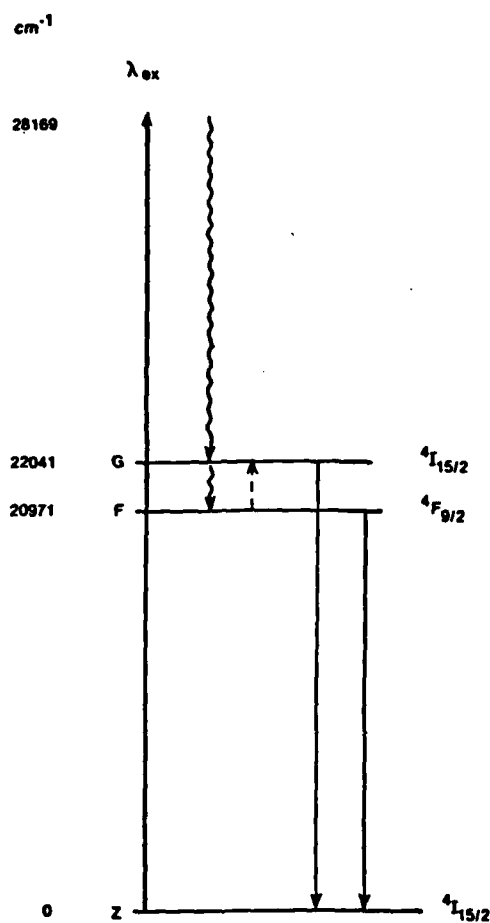
Figure 2. Energy Levels of Dy^{+3}:LaF_3 Involved in Thermalization Process.

is displayed. Notice the gradual build-up of the G-fluorescence line located at 4537 Å with temperature.   The G level increases in intensity approximately 200 fold over a 973-K temperature range. In the case of thermalization, the variation of the upper population and, thus, intensity is expressed by the Boltzmann equation
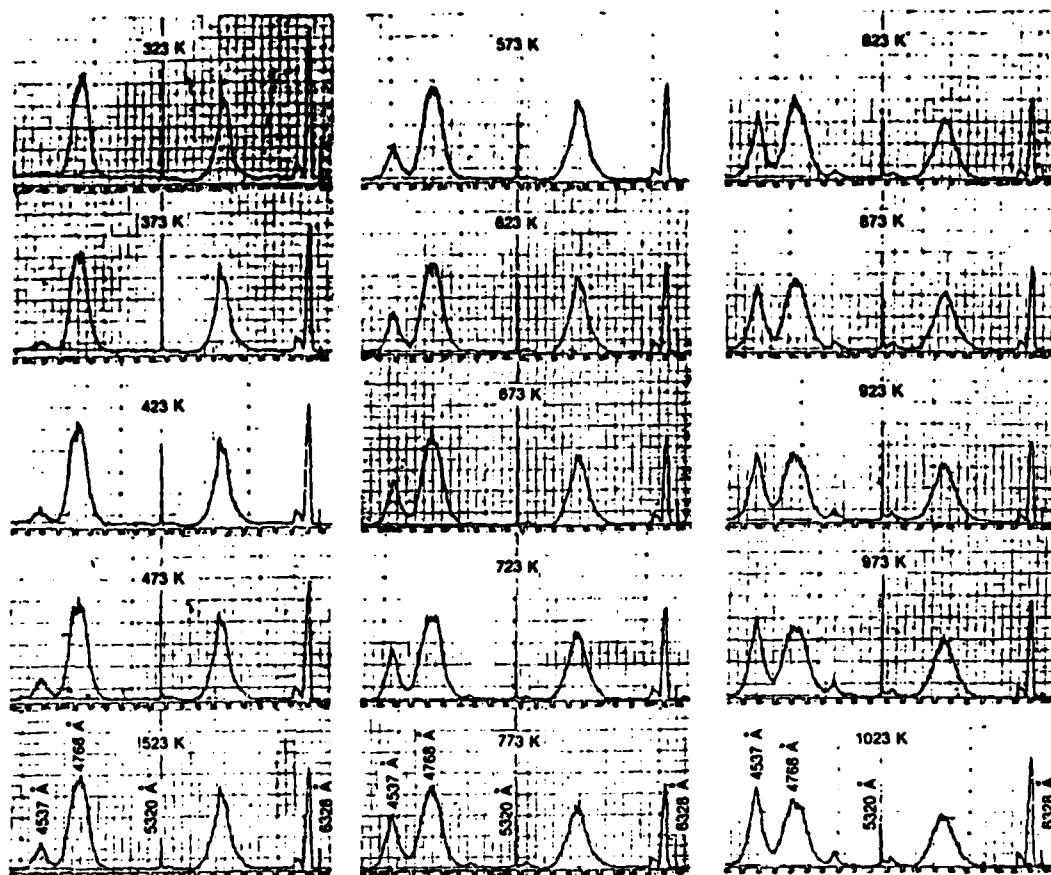
$$I(T) = I_0 \exp(-\Delta E/kT) \tag{1}$$

Figure 3. Spectral Variation of Dy^{+3}:LaF_3 with Temperature.

where ΔE is the energy gap between the two levels (~ 1070 cm^{-1}), T the temperature in degrees Kelvin, k the Boltzmann constant, and I(T) the relative changes in intensity of the upper fluorescence level. Figure 4 is a plot of the experimental data for Dy^{+3}. The solid line represents results obtained with Eq. (1), assuming a 1070 cm^{-1} energy gap. As depicted in this figure, the agreement with the Boltzmann equation is quite good. The agreement with Eq. (1) substantiates the occurrence of a thermalization process.
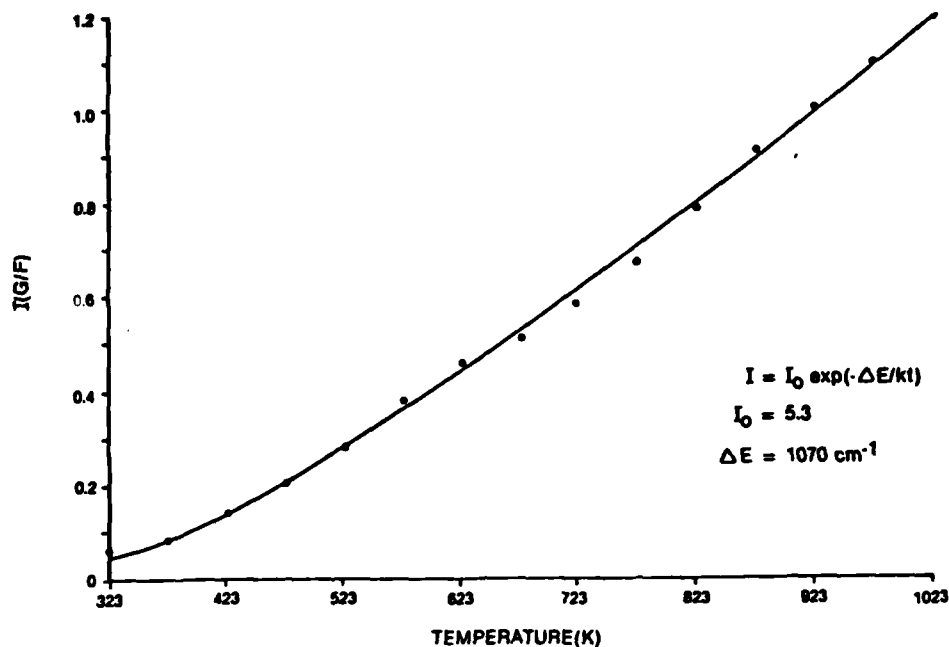
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Figure 4. Experimentally Observed Variation of the G to F Fluorescence Levels of Dy^{+3}:LaF_3 with Temperature. Solid line represents fit to data, assuming energy gap of 1070 cm^{-1}.

An important feature of note concerning the Dy^{+3}:LaF_3 fluorescence is that the adjacent F-level peak at 4768 Å displays very little change with temperature and, thus, can be used as an internal standard for calibration of the G level. This is important when working in adverse environments where an absolute-intensity measurement of a single line is imposible but a relative measurement to a nearby line is practical. Thus, the G-F fluorescence of Dy^{+3}:LaF_3 promises to be extremely useful for surface temperature measurements.

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V. LIFETIME MEASUREMENTS

To understand the effect of temperature on the lifetimes of rare-earth transitions, one must examine the effect of temperature on the radiative and nonradiative relaxation rates which govern the observed lifetimes. The lifetime τ_a of an excited state a is given by[9]

$$\frac{1}{\tau_a} = \sum_b W_{ab}^R + \sum_b W_{ab}^{NR}$$

where the summations are for transitions terminating on all final states b. The radiative probability W^R includes both purely electronic and phonon-assisted transitions; the nonradiative probability W^{NR} includes relaxations by multi-phonon emission and effective energy-transfer rates arising from ion-ion interactions. Radiative decay rates can range from 10 to 10^6/s but are not temperature dependent. The temperature dependence of the lifetime is thus solely determined by the temperature dependence of the nonradiative relaxation rate.

Nonradiative relaxation between J states can occur by the simultaneous emission of several phonons which are sufficient to conserve the energy of the transition. These multi-phonon processes arise from the interaction of the rare-earth ion with the fluctuating crystalline electric field. The crystal field at the ion site is not static but subject to oscillatory behavior due to the vibrations of the lattice or molecular groups. The

lattice vibrations are quantized as phonons having symmetry properties determined by the symmetry of the crystal and excitation energies determined by the masses of the constituent ions and the binding forces.

Consider a multi-phonon relaxation across an energy gap ΔE to the next-lowest level. The number of phonons p_i of equal energy $\hbar\omega_i$ required to conserve energy and, hence, the order of the process is determined by the condition

$$p_i \hbar\omega_i = \Delta E$$

The temperature dependence is

$$W(T) = W_0 (n_i + 1)^{p_i}$$

where n_i is the occupation number of the ith phonon mode and W_0 is the spontaneous transition rate. Replacing n_i by its Bose-Einstein average,

$$n_i = [\exp(\hbar\omega_i/kT) - 1]^{-1}$$

The temperature-dependent multi-phonon transition rate for a single-frequency p-phonon process is

$$W^{p_i}(T) = W_0 \left[\frac{\exp(\hbar\omega_i/kT)}{\exp(\hbar\omega_i/kT)^{-1}} \right]^{-1} \tag{2}$$

From Eq. (2) it can be seen that the order (number of phonons required for energy conservation) of the process is the critical feature in the temperature dependence. Figure 5 shows the relative transition rates for a fourth-, fifth-, and sixth-order process.[10] It should be noted that the higher the order of the multi-phonon process, the higher the nonradiative
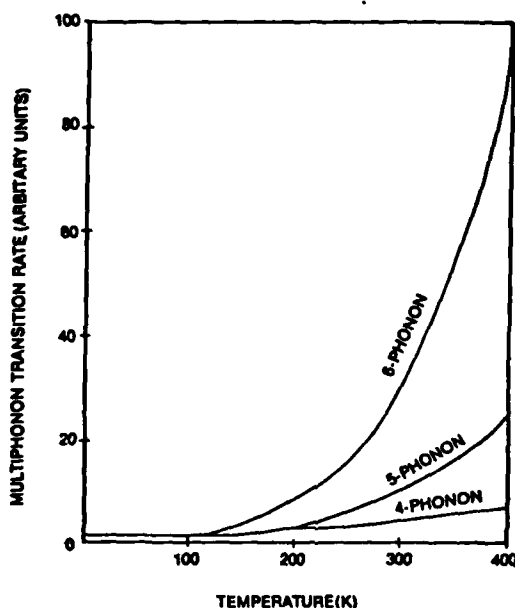

Figure 5. Variation of Multi-phonon Relaxation Rate with
Temperature and Phonon Number (From Ref. 10).

transition rate, which translates into a shorter lifetime for the radiative process. In actuality, the single-frequency phonon model [the result of which is given in Eq. (2)], is limited since decay occurs between Stark levels of two J multiplets. Since the levels within the multiplet are in thermal equilibrium, the combined rate is a Boltzmann average of the rates from the separate levels given by

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$$W = \frac{\sum_a \sum_b g_a W_{ab}(T) \exp(-\Delta_a/kT)}{\sum_a g_a \exp(-\Delta_a/kT)} \qquad (3)$$

where W_{ab} is an individual decay rate from an upper multiplet level to a lower multiplet level b, g_a is degeneracy, and Δ_a the energy separation of the ath level from the bottom level of the upper multiplet. Equation (3) is the precise description of the characteristics of the temperature dependence; however, of most value is establishing the order of the multi-phonon decay process and the energies of the dominant phonons involved. Experimental studies of the temperature dependence confirm that the order of the process is often the lowest order consistent with energy conservation and the cutoff frequency of the phonon spectrum.[7,11]

The classic example of the effect of temperature on the lifetime of a transition is given by the R-fluorescence lines of ruby ($Cr^{+3}:Al_2O_3$). Because of the importance of ruby as a laser source, this material has been widely studied. The lifetime dependence on temperature has been noted by several investigators and was first discussed for temperatures above 300 K by Nelson.[12] In this work it was noted that the fluorescence lifetimes of the R lines decrease by a factor of two from 273 to 373 K. A temperature probe based upon the measurement of the R-line lifetimes was reported in Ref. 13.

Ruby was chosen as a candidate for study because of its high sensitivity to temperature; however, its lifetimes above 400 K have not been studied in detail. The experimentally observed lifetimes as measured by a Model 1800

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$^4A_2$ state. Thus, in order to nonradiatively relax this state, a high-order multi-phonon process would be required. As mentioned above, the higher the phonon number, the greater the temperature effect on lifetimes and, thus, the more sensitive the transition to temperature change. In this case the phonon number was determined to be 9; thus, as demonstrated in Fig. 5, the multi-phonon rate would be expected to increase greatly with temperature, resulting in a sharp decrease in the fluorescence lifetime. Notice that the lifetime of the R-fluorescence lines drops below 10 μs at 800 K, which is approximately the cutoff for an accurate measurement of the lifetime with the Model 1800 Biomation. This cutoff would allow one to discriminate against fluorescence not emitted from the solid surface because of the extremely steep temperature gradient just above the surface.

Another lifetime system which displays a lower-order phonon process is $Er^{+3}:CaF_2$. The energy level of interest in this rare-earth ion is the $E(^4S_{3/2})$ level which is separated from the next-lowest-lying level by 2700 cm^{-1}. The cutoff for the phonon spectrum of CaF_2 is $\sim$ 500 cm^{-1}; thus, the phonon order of the relaxation of this level is $\sim$ 5. As displayed in Fig. 7, a fifth-order process is less sensitive to temperature changes and, thus, covers a much wider temperature range. As indicated in Fig. 7, the relaxation process is still detectable beyond 1000 K, allowing an extension to higher temperatures when required.
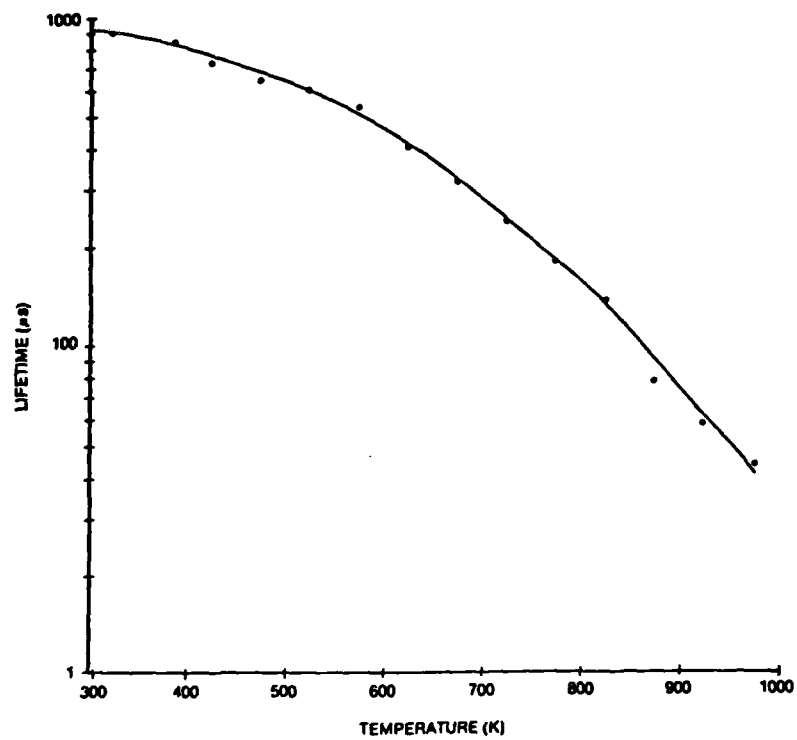
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Figure 7. Experimentally Measured Lifetimes of the
5374 Å Fluorescence Level of $Er^{+3}:CaF_2$ as
Function of Temperature.

VI. CONCLUSIONS

In conclusion three rare-earth-ion crystals have been identified as good candidates for surface-temperature measurements on energetic materials. The G and F levels of Dy^{+3}:LaF_3 displayed a thermalization process which was quite sensitive to temperature in the 300-1000 K range because of the 1070 cm^{-1} energy gap of these levels. The lifetime of the R fluorescence line of Cr^{+3}:Al_2O_3 (ruby) displays an extreme sensitivity to temperature through a multi-phonon relaxation of the radiative transition. The R fluorescence displayed a 3.4-ms lifetime at 300 K which decreases to 15 μs at 773 K. The 5374-Å fluorescence line of Er^{+3}:CaF_3 is nonradiatively relaxed through a fifth-order multi-phonon process, and its lifetime displays a high sensitivity to temperature. The lifetime sensitivity of the 5374-Å line is lower than that of the R line of ruby but covers a wider temperature range.

VII. PRESENTATIONS DURING THIS REPORTING PERIOD

The following presentations were made during this reporting period:

"Application of Atomic Fluorescence to Measurement of Combustion Temperature in Solid Propellants" (L. P. Goss and A. A. Smith), Presented by L. P. Goss at the 1984 AFOSR Research Meeting on Diagnostics of Reacting Flows, March 21-22, 1984, New Haven, CT.

"Application of Atomic Fluorescence to Measurement of Surface Temperature in Solid Propellants" (L. P. Goss and A. A. Smith), Presented by L. P. Goss at the 21st JANAAF Combustion Meeting, October 1-5, 1984, Laurel, MD (to be published in the conference proceedings).

"Laser-Induced Fluorescence as a Measurement of Surface Temperature in Solid-Fuel Propellants" (L. P. Goss and A. A. Smith), Presented by A. A. Smith at Lasers '84, November 26-30, 1984, San Francisco, CA (to be published in the conference proceedings).

The authors received awards from SRL's Board of Awards for these presentations.

VIII. ANTICIPATED EFFORT

Work is continuing on LaF_3:0.5%Ho and LaF_3:0.5%Er--newly acquired crystals which are yielding positive results in both lifetime and line-strength measurements. As other crystals are found to be acceptable, they will be included in the fast-pyrometry tests to follow in Phase II.

The next step in Phase II involves verification of temperature measurements by way of high-speed pyrometry. Equipment has been received to begin construction of a device to permit these measurements.

Future work will involve embedding species into model energetic materials and comparing temperature data obtained from LIF measurements to those obtained through measurements by more conventional pyrometry methods.

Respectfully submitted,

SYSTEMS RESEARCH LABORATORIES, INC.

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